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REQUEST FOR CONTINUED EXAMINATION (RCE) TRANSMITTAL

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Application Number	10/073,755
Filing Date	02/11/02
First Named Inventor	Black et al.
Art Unit	2862
Examiner Name	S. Zaveri
Attorney Docket Number	YOR920010467US1

This is a Request for Continued Examination (RCE) under 37 CFR 1.114 of the above-identified application.
Request for Continued Examination (RCE) practice under 37 CFR 1.114 does not apply to any utility or plant application filed prior to June 8, 1995, or to any design application. See Instruction Sheet for RCEs (not to be submitted to the USPTO) on page 2.

1. Submission required under 37 CFR 1.114 Note: If the RCE is proper, any previously filed unentered and amendments enclosed with the RCE will be entered in the order in which they were filed unless applicant instructs otherwise. If applicant does not wish to have any previously filed unentered amendment(s) entered, applicant must request non-entry of such

a. ☐ Previously submitted. If a final Office action is outstanding, any amendments filed after the final Office action may be considered as a submission even if this box is not checked.
   i. ☐ Consider the arguments in the Appeal Brief or Reply Brief previously filed on ______
   ii. ☐ Other ______

b. ☒ Enclosed
   i. ☐ Amendment/Reply
   ii. ☐ Affidavit(s)/Declaration(s)
   iii. ☒ Information Disclosure Statement (IDS)
   iv. ☐ Other ______

2. Miscellaneous
   a. ☐ Suspension of action on the above-identified application is requested under 37 CFR 1.103(c) for a period of ______ months. (Period of suspension shall not exceed 3 months; Fee under 37 CFR 1.17(i) required)
   b. ☐ Other

3. Fees The RCE fee under 37 CFR 1.17(e) is required by 37 CFR 1.114 when the RCE is filed.
   a. ☒ The Director is hereby authorized to charge the following fees, or credit any overpayments, to Deposit Account No. 50-0510
      i. ☒ RCE fee required under 37 CFR 1.17(e)
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Name (Print / Type)	Mohammad S. Rahman	Registration No. (Attorney / Agent)	43,029
Signature	Mohammad S. Rahman	Date	10-14-2003

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In re patent application of

Black et al.

Serial No.: 10/073,755

Group Art Unit:2862

Filing Date: February 11, 2002

Examiner: S. Zaveri

For: MAGNETIC-FIELD SENSOR HAVING MAGNETIC NANOPARTICLES

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INFORMATION DISCLOSURE STATEMENT

Sir:

Under the provisions of 37 CFR §1.97 through §1.99 and pursuant to applicants' duty of disclosure under 37 CFR §1.56, applicants respectfully bring the following documents listed on the attached form PTO-1449, to the attention of the Examiner in charge of the above-identified application. A copy of the listed document is provided herewith for the convenience of the Examiner. This citation does not constitute an admission that the references are relevant or material to the claims. They are only cited as constituting related art of which the applicants are aware.

It is respectfully requested that the listed references be considered by the Examiner and formally made of record in this application.

Please charge any deficiencies in fees and credit any overpayment of fees to Attorney's Deposit Account No. 50-0510.

Respectfully submitted,

Mohammad S. Rahman

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Registration No. 43,029

Date: 10/14/2003

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INFORMATION DISCLOSURE CITATION

(Use several sheets if necessary)

Docket Number (Optional)	Application Number
YOR920010467US1	10/073,755
Applicant(s)	
Black et al.	
Filing Date	Group Art Unit
02/11/02	2862

U.S. PATENT DOCUMENTS

*EXAMINER INITIAL	REF	DOCUMENT NUMBER	DATE	NAME	CLASS	SUBCLASS	FILING DATE IF APPROPRIATE

FOREIGN PATENT DOCUMENTS

	REF	DOCUMENT NUMBER	DATE	COUNTRY	CLASS	SUBCLASS	Translation YES	Translation NO

OTHER DOCUMENTS *(Including Author, Title, Date, Pertinent Pages, Etc.)*

		K. Nakajima et al., "Magnetoresistance Oscillations in Double Ferromagnetic Tunnel Junctions with Layered Ferromagnetic Nanoparticles", IEEE Transactions on Magnetics, Vol. 36, No. 5, 2000, pp. 2806-2808.

EXAMINER	DATE CONSIDERED

EXAMINER: Initial if citation considered, whether or not citation is in conformance with MPEP Section 609; Draw line through citation if not in conformance and not considered. Include copy of this form with next communication to applicant.

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Magnetoresistance Oscillations in Double Ferromagnetic Tunnel Junctions with Layered Ferromagnetic Nanoparticles

K. Nakajima, Y. Saito, S. Nakamura, and K. Inomata

Abstract—Tunnel magnetoresistance in double tunnel junctions with layered ferromagnetic nanoparticles was investigated. The sample comprises two ferromagnetic electrodes (CoFe/Fe) separated by an Al_2O_3 insulating layer in which layered $Co_{80}Pt_{20}$ nanoparticles are embedded. The nanoparticles are ellipsoidal with an average diameter of 3.7 nm, and composes a well-defined layer. At 10 K, we observed magnetoresistance oscillation with respect to the bias voltage. The observed TMR oscillation, with a period of 1.6 mV, accompanied oscillations of the conductance. We consider that the conductance oscillation may originate from the single electron charging effect.

Index Terms—Single electron tunneling effect, double tunnel junction, nanoparticle, tunnel magnetoresistance.

I. Introduction

MAGNETORESISTANCE (MR) phenomena in nano-scale ferromagnetic tunnel junctions currently attract much attention. When the capacitance C of the junction is so small that the charging energy $E_c = e^2/2C$ can be larger than the thermal energy k_BT, single electron tunneling (SET) effects play an important role. The basic system in which the SET effects such as the Coulomb blockade and the Coulomb staircase occur is small "center island" connecting to macroscopic reservoirs through tunnel barriers. Often the center island is coupled to a gate capacitor with a gate voltage V_G, namely this is the SET transistor. The SET effects and the SET transistor have already been extensively investigated. Recently, Ono et al. succeeded in fabricating the ferromagnetic SET transistor, and clearly observed the enhancement of the tunnel magnetoresistance (TMR) in the Coulomb blockade regime and the magneto-Coulomb oscillations [1]. Their studies attracted considerable attention to the interplay of the spin-dependent electron transport and the SET effect. Aside from the ferromagnetic SET transistor, the spin-polarized SET effects were also anticipated in more complicated systems, such as ferromagnetic granular films with current-in-plane geometry [2], [3], granular films with current-perpendicular-to-plane geometry using STM tips [4], or small point contacts [5], [6], and hybrid-type ferromagnetic double tunnel junctions (DTJ's) where layered ferromagnetic nanoparticles are embedded in the tunnel barrier [7]–[9].

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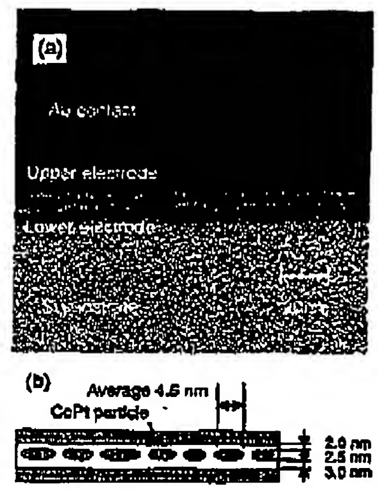

Fig. 1. Transmission electron micrograph of the cross section of the sample. (b) Schematic of the sample cross section.

In this paper, we focus on the hybrid-type ferromagnetic DTJ's. We report unique behavior of TMR observed in the ferromagnetic DTJ's in which layered ferromagnetic $Co_{80}Pt_{20}$ nanoparticles were embedded.

II. Experimental

The DTJ's were prepared onto Si(001) substrates by ion beam sputtering and conventional magnetron sputtering technique. An Fe 6 nm /Co_1Fe_1 3 nm bilayer and a Co_1Fe_1 6 nm/Fe 12 nm/Au 98 nm trilayer were used as the bottom and the top electrode, respectively. The double tunnel barrier with structure, $Al_2O_3/Co_{80}Pt_{20}/Al_2O_3$, was prepared by alternatively sputtering from Al_2O_3 and $Co_{80}Pt_{20}$ targets. The higher surface energy of $Co_{80}Pt_{20}$ compared to that of Al_2O_3 leads to 3-dimentional nucleation of the deposit. Photolithography and Ar etching technique formed the cross pattern structures. The junction area was 80×80 μm^2. The layered structure was investigated by cross-sectional transmission electron microscopy (TEM). The I–V and the MR curves were measured by a dc-fourterminal method as a function of the bias voltage and of the magnetic field.

III. Results and Discussion

Fig. 1(a) shows a cross-sectional TEM image of a device with a thickness, $Co_{80}Pt_{20}$ 1.9 nm. The gray portions pinched by the

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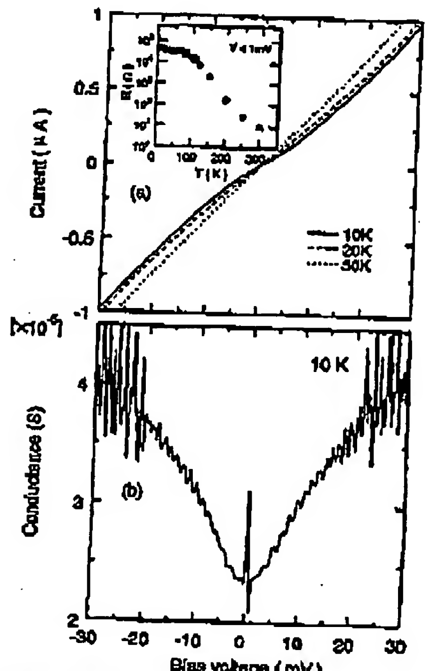

Fig. 2. (a) Current-voltage characteristics at various temperatures. Inset shows the temperature dependence of low-voltage tunnel resistance measured below 1 mV. (b) Conductance-voltage characteristic.

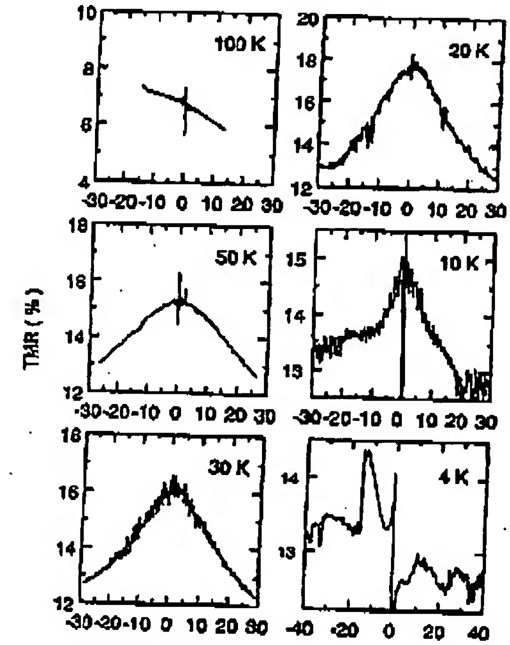

Fig. 3. Bias voltage dependence of the tunnel magnetoresistance at various temperatures.

electrodes are the Al_2O_3 tunnel barriers. The thicknesses of the top and bottom barrier layer are approximately 2 and 3 nm, respectively. The $Co_{80}Pt_{20}$ nanoparticles arranges in a well-defined layer between the double tunnel barriers. A schematic illustration of the sample cross section is shown in Fig. 1(b). The particles are ellipsoidal with a height of 2.5 nm, and its width is distributed at around 4.5 nm. The half-width of the distribution is about 1 nm. Assuming spherical shaped particles, we get an average diameter of 3.7 nm, and can roughly estimate E_c^0 of an individual particle to be about 50 meV. The feature of this system is that the barrier thickness s is constant regardless of the size of the granules d, unlike the granular films where ratio s/d keeps constant [2]. This system is thus considered to be a parallel connection of nanoscale DTJ's in which a ferromagnetic nanoparticle plays as a "center island".

Fig. 2(a) shows the I–V curves of the sample measured at 10, 20 and 50 K. Also shown is the temperature dependence of the low-voltage tunnel resistance. At room temperature (RT), the tunnel resistance is below 10 Ω, however, as temperature decreases, it increases exponentially, and exceeds the quantum resistance at 50 K. At 10 K, nonlinearity of I–V curves become more remarkable, and we observe a distinct oscillation on the conductance data in Fig. 2(b). The conductance is calculated from the I–V curve using the trapezoid formula. The conductance oscillation attains to twenty or more with a period of 1.6 mV, indicating the appearance of Coulomb staircase. The observed period is a tenth or less of the charging energy E_c^0 of an individual particle. We consider that the lower value observed may originate from the sample structure, the parallel connection of DTJ's. The I–V characteristic of the parallel connection will be mostly decided by DTJ's with higher conductance, namely, with larger particle and lower charging energy. Hence, it is considered that the effect of the size-distribution of the particles may lower the effective value of the charging energy. Beside, it is necessary to consider the capacitive couplings between the particles that may reduce the effective value of the charging energy.

At RT, the MR curves showed superparamagnetic behavior of the nanoparticles. Below 200 K, the curves showed coercivities, and zero-bias TMR steeply increased around 100 K from few % to 15% at 50 K. Fig. 3 shows the bias voltage dependence of the TMR measured at various temperatures. The bias voltage dependence of the TMR was calculated from the two I–V curves measured for both parallel and anti-parallel configurations between the nanoparticles and the electrode magnetizations. Above 20 K, the TMR shows a broad peak at the zero voltage, and monotonously decreases with the increase in voltage. The decrease is essential to the spin-polarized tunneling effects irrespective of the SET effects. However, when the sample is cooled below 20 K, the bias voltage dependence dramatically changes. At 10 K, one sees a clear oscillation with a period of 1.6 mV, which is identical to that observed in the conductance oscillation. We consider that the TMR oscillation originates from the conductance oscillation, as a result of the SET effects. At 4 K the oscillating behavior becomes more prominent. The TMR largely oscillates with a period of 16 mV, and consequently, the zero-bias peak disappears completely. At relatively high bias region, one can see that another oscillation with the same period observed at 10 K superposes on the large oscillation. However, unlike 10 K, conductance oscillations corresponding to the large oscillation were not clearly observed. Moreover, it is somewhat strange that

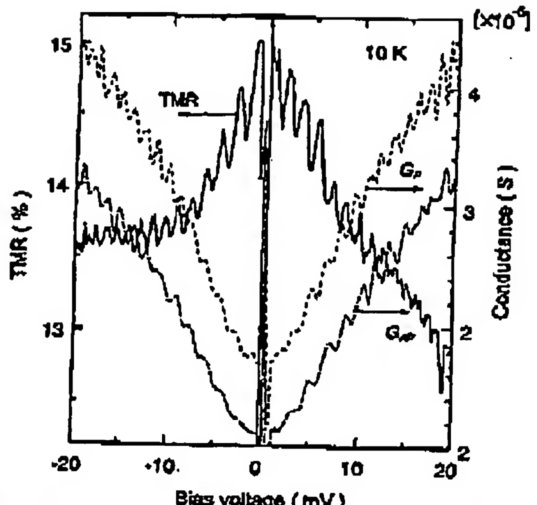

Fig. 4. Bias voltage dependence of the tunnel magnetoresistance and the tunnel conductance measure at 10 K.

the large oscillation follows the short oscillation. Here, we can give no definite conclusion whether the large osculation also originates from the SET's effect or not. More studies such as the particle size dependence of the oscillations period are now in progress.

Fig. 4 shows the bias voltage dependence of the TMR and the tunnel conductance G_P and G_{AP} measured at 10 K. G_P and G_{AP} denote the conductances for parallel and anti-parallel configurations between the nanoparticles and the electrode magnetizations, respectively. Note the observed TMR oscillation accompanies oscillations of both G_P and G_{AP}. Furthermore, one sees the positions of the conductance maxima are slightly different with respect to the relative orientation of the magnetization between the electrodes and the nanoparticles. Within the limit of fast spin-flip relaxation time, the threshold voltage in the Coulomb staircase is identical to both parallel and anti-parallel configuration. Under this limitation, Majunder and Hershfield calculated the TMR for ferromagnetic DTJ's in the Coulomb blockade regime, and found spikes in the bias dependence of the TMR, which occur at the threshold voltage in the Coulomb staircase where the current increases by steps [10]. On the contrary, very recently, several groups have presented generalizations of the orthodox theory to describe the nonequilibrium spin polarized states in the "center island" of the ferromagnetic SET transistor by introducing the appropriate spin-flip relaxation time [11], [12]. At the regime where the spin-flip relaxation time is much longer than the tunneling time, owing to the nonequilibrium spin accumulation spin splitting of the threshold voltage occurs [11], [13]. We believe that these out of phase conductance oscillations indicate a possibility of observing the nonequilibrium spin accumulation.

It is what should be surprised that even though macroscopic electrodes were used, the SET effects could be observed. It is considered that existence of the size distribution of the particles may smear out the SET effects. If we assume a size distribution of 20%, a distribution of the charging energy amounts to 30%. We consider the relatively small size distribution of the particles may be one of the reasons for successful observation. As stated previously, the structure of our sample, the parallel connection of the DTJ's, may also be committed advantageously, because most of the DTJ's in the parallel connection become blocked at low temperature.

IV. Conclusion

We have successfully fabricated double tunnel junctions comprised two ferromagnetic electrodes (CoFe/Fe) separated by an Al_2O_3 insulating layer in which layered $Co_{80}Pt_{20}$ nanoparticles were embedded. At 10 K, we observed magnetoresistance oscillation with respect to the bias voltage. The observed TMR oscillations, with a period of 1.6 mV, accompanied oscillations of the conductance, indicating the discrete electrostatic potential in the nano particles. The positions of the conductance maxima are slightly different with respect to the relative orientation between the particles and the electrodes magnetizations. We believe that these out of phase conductance oscillations indicate the possibility of observing the nonequilibrium spin accumulation.

Acknowledgment

The authors are grateful to Prof. J. Inoue, Nagoya University for helpful discussions.

References

[1] K. Ono, H. Shimada, and Y. Ootsuka, "Magnetoresistance of Ni/NiO/Co small junctions in Coulomb blockade regime," *J. Phys. Soc. Jpn.*, vol. 65, pp. 3449–3451, Nov. 1997.

[2] J. S. Helman and B. Abeles, "Tunneling of spin-polarized electrons and magnetoresistance in granular Ni films," *Phys. Rev. Lett.*, vol. 37, pp. 1429–1432, 1976.

[3] S. Mitani, S. Takahashi, K. Yakushiji, S. Maekawa, and H. Fujimori, "Enhanced magnetoresistance in insulating granular systems: Evidence for higher-order tunneling," *Phys. Rev. Lett.*, vol. 81, pp. 2799–2802, 1998.

[4] K. Takanashi, S. Mitani, J. Chiba, and H. Fujimori, Scanning tunneling microscopy investigation of single electron tunneling in Co-Al-O and Cu-Al-O granular films, in *J. Appl. Phys.*, submitted for publication.

[5] R. Desmicht, G. Faini, I.-V. Cros, A. Fert, F. Petroff, and A. Vaures, "Point-contact electrodes to probe charging effects in individual ultrasmall cobalt clusters," *Appl. Phys. Lett.*, vol. 72, pp. 386–388, 1998.

[6] S. Gueron, M. M. Deshmukh, E. B. Myers, and D. C. Ralph, "Tunneling via individual electronic states in ferromagnetic nanoparticles," *Phys. Rev. Lett.*, vol. 83, pp. 4148–4151, 1999.

[7] K. Inomata, H. Ogiwara, Y. Saito, K. Yusu, and K. Ichihara, "Spin-dependent tunneling between a soft ferromagnetic layer and hard magnetic nanoparticles," *Jpn. J. Appl. Phys.*, vol. 36, pp. L1380–L1383, 1997.

[8] K. Inomata and Y. Saito, "Spin-dependent tunneling through layered ferromagnetic nanoparticles," *Appl. Phys. Lett.*, vol. 73, pp. 1143–1145, 1998.

[9] L. F. Schelp, A. Fert, F. Fettar, P. Holody, S. F. Lee, J. L. Maurice, F. Petroff, and A. Vaures, "Spin-dependent tunneling with Coulomb blockade," *Phys. Rev.*, vol. B56, pp. R5747–R5750, 1997.

[10] K. Majumder and S. Hershfield, "Magnetoresistance of the double-tunnel-junction Coulomb blockade with magnetic metals," *Phys. Rev.*, vol. B57, pp. 11 521–11 525, 1998.

[11] J. Barnas and A. Fert, "Interplay of spin accumulation and Coulomb blockade in double ferromagnetic junctions," *J. Magn. Magn. Mater.*, vol. 192, pp. L391–L395, 1999.

[12] A. Brataas, Y. N. Nazarov, J. Inoue, and G. E. Bauer, "Spin accumulation in small ferromagnetic double tunnel junctions," *Phys. Rev.*, vol. B59, pp. 93–96, 1999.

[13] A. Brataas, M. Hirano, J. Inoue, and G. E. W. Bauer, private communication.